Complexity of Grain Boundary Motion: Insights from Molecular Dynamics

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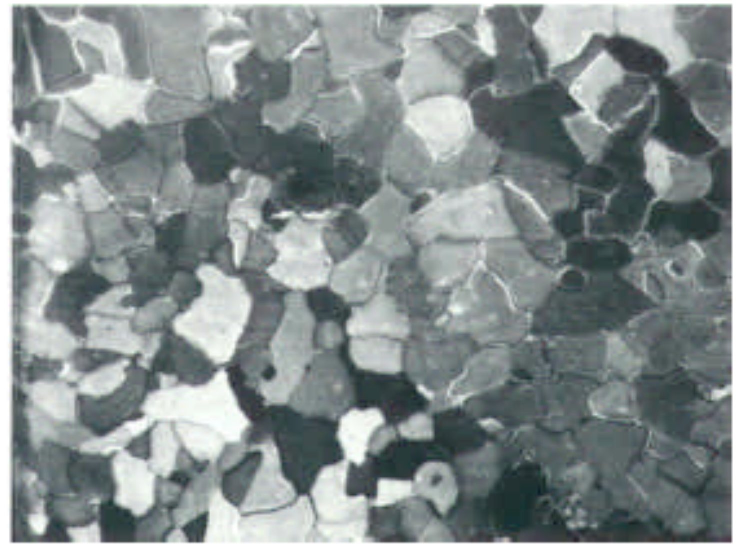
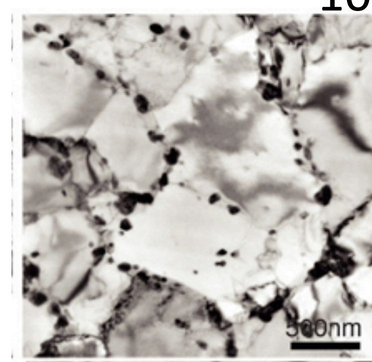
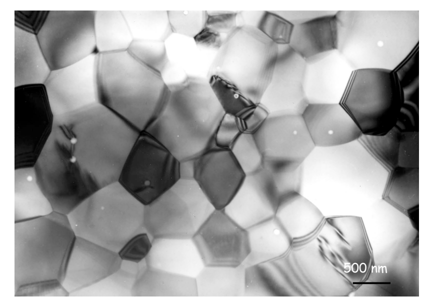
Why does anyone care about grain growth?

- Grain-level microstructure strongly influences a wide range of materials properties
  - Strength
    - Hall-Petch relationship: $\sigma_y = \sigma_0 + \frac{k_y}{\sqrt{d}}$
  - Toughness and Fracture
  - Corrosion resistance
  - Electrical conductivity
  - Magnetic susceptibility
  - …
- *Controlling the microstructure and relating the microstructure to properties are central problems in materials science.*



1000 µm



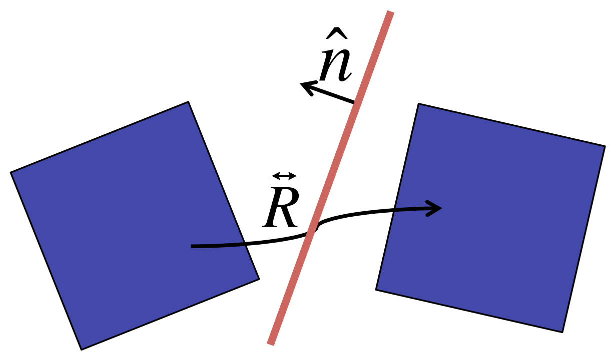


Evolution of grain microstructure is a highly complex multi-scale modeling problem

- Atomic-scale
  - Boundary properties are determined by atomic-scale structure and dynamics
    - Energy – changes in *atomic-level* bonding/coordination in the boundary
    - Motion – local *atomic-level* rearrangements at the boundary
  - Time-scale: picoseconds – nanoseconds
- Meso-scale
  - Grain sizes: ~10 nanometers - ~100 micrometers
  - Need to consider the 3-D network of grain boundaries
  - Time-scale: seconds to hours
- Conventional strategy
  - Determine the properties of grain boundaries with atomic-scale methods
  - Evolve the grain structure with meso-scale simulations that incorporate the boundary properties – energy, mobility

What is the big deal about determining grain boundary properties?

- *"We hold these truths to be self-evident, that all grain boundaries are **NOT** created equal, …"* - apologies to Thomas Jefferson
  - There is a **5-dimensional** space of macroscopic grain boundary structure
  - The properties vary throughout this 5-D space in an, at best, partially understood manner
  - And this does ***NOT*** even consider the effects of temperature, alloying, impurities, second phases, applied stress, …
  - For a given macroscopic configuration, multiple microscopic (atomic-level) grain boundary structures may be present in equilibrium



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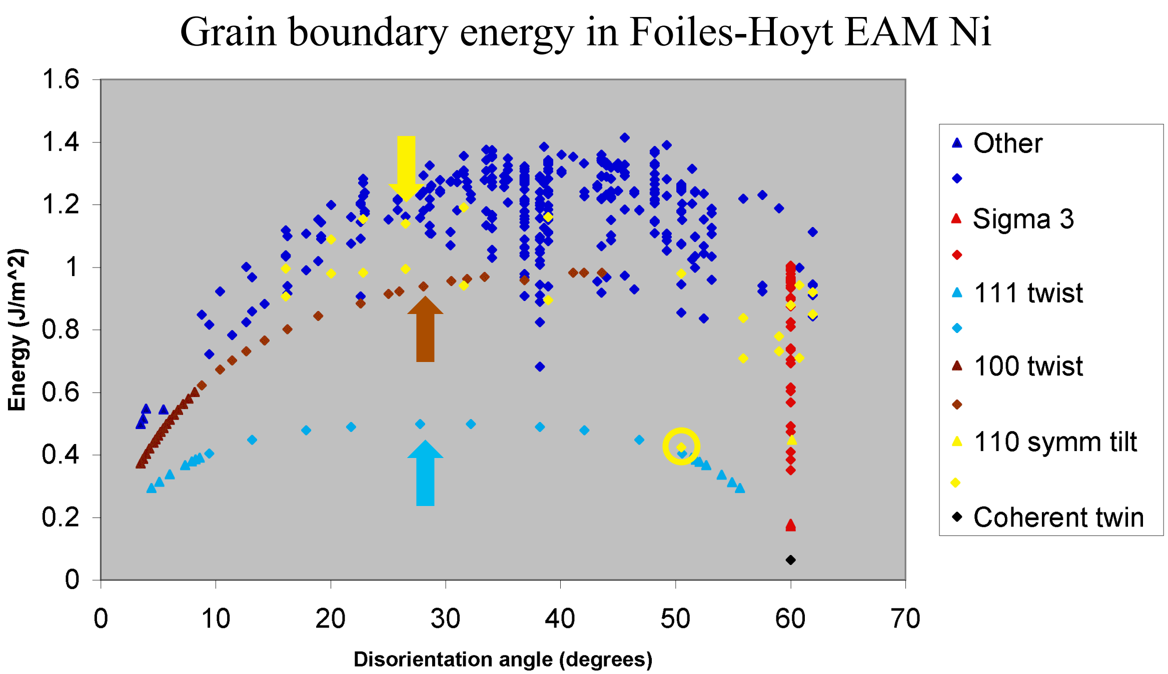
Grain Boundary Properties have been computed for a large (388 sample) catalog of boundaries

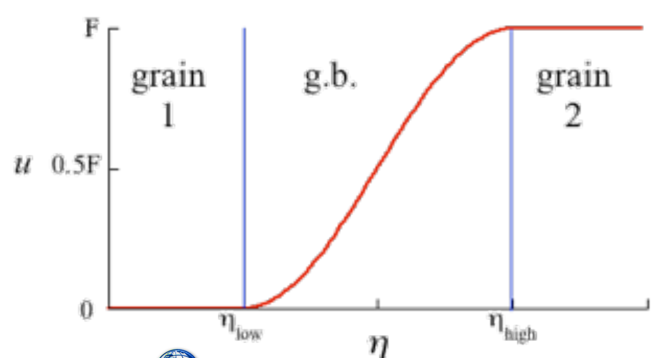


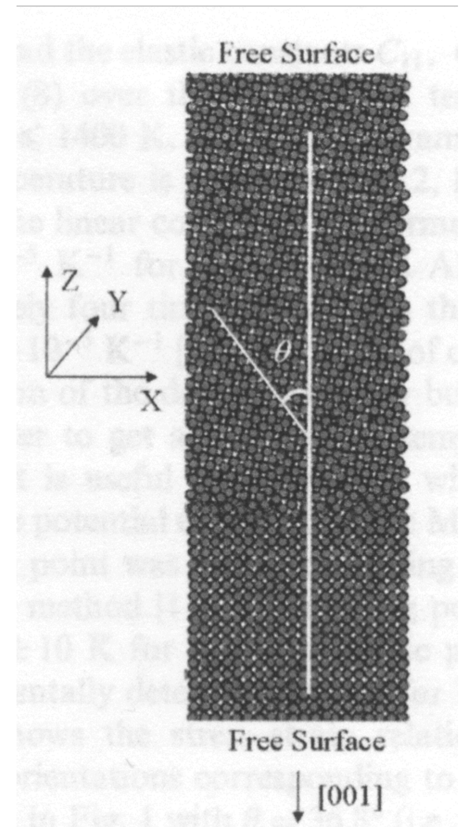
Misorientation is insufficient to determine grain boundary energy

- <111> twist boundaries are lowest in energy and obey Read-Shockley theory
- <100> twist boundaries are also low energy and obey Read-Shockley
- The $\Sigma11$ symmetric <110> tilt boundary with [311] planes is anomalously low in energy
- Other symmetric <110> tilt boundaries vary widely
- Large spread in energy for the $\Sigma3$ grain boundaries
- The spread of "random" boundary energies is large - almost a factor of 2

Grain boundary mobility has been determined by the application of an external driving force

- Possible external driving forces
  - Strain
    - Relies on elastic anisotropy which produces differing strain energy densities in the two crystals
    - Not applicable for all boundaries
  - Synthetic Driving Forces
    - Artificial potential and corresponding forces added to the system which favors growth of one grain
- The magnitude of the driving force is a method parameter
  - Important to find the low driving force limit





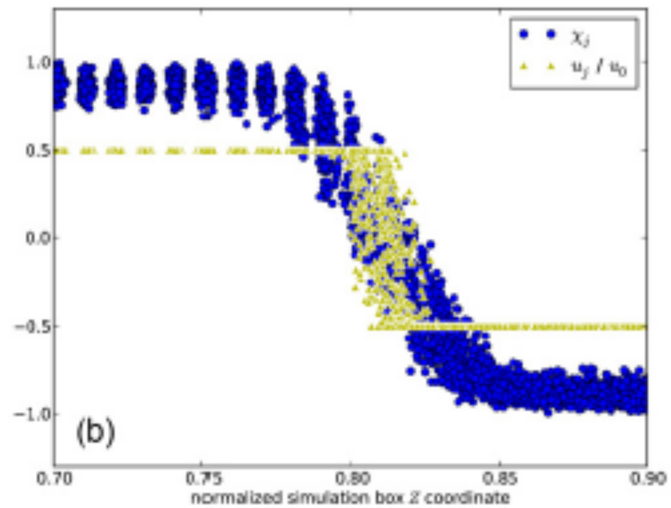
Zhang, Mendelev & Srolovitz, Acta Mater. **52**, 2569 (2004)

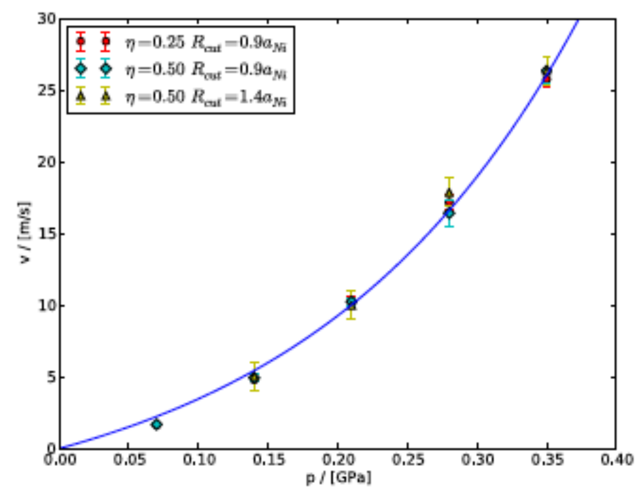
Synthetic driving force has been developed that conserves energy

- Discontinuity-free orientation measure based on local diffraction condition

$$\chi_j = \frac{1}{N}\left[\sum_\alpha \left|\sum_k w(|\vec{R}_{jk}|)e^{i\vec{Q}_\alpha \vec{R}_{jk}}\right|^2 - \sum_\beta \left|\sum_k w(|\vec{R}_{jk}|)e^{i\vec{Q}_\beta \vec{R}_{jk}}\right|^2\right]$$

  - Q are reciprocal lattice vectors for desired grain orientations
  - w(R) is a localization function
- Features of new approach
  - No discontinuities – excellent energy conservation
  - Thermodynamic intergration for free energy density difference can be avoided in many cases
  - Can tune the degree of locality





Ulomek, O'Brien, Foiles, Mohles, MSMSE 23 (2015), 025007.

Mobility methods have been compared for a specific boundary and potential

- Reference grain boundary and potential
  - Σ5[0 1 0] asymmetric tilt boundary with (1 0 7) and (1 0 1) boundary planes
  - Ackland et al. EAM potential for Ni
- 3 methods yield values for mobility that agree within error bars
  - No driving force (fluctuation)
    - Adapted Interface Random Walk
  - Driving Force (low force limit)
    - Elastic Strain
    - Synthetic Driving force
      - Ulomek, et al, MSMSE 23 (2015) 025007

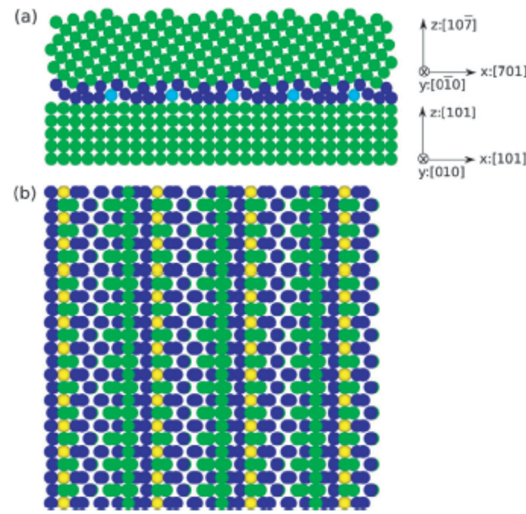


Figure 1. (*a*) The Atomistic configuration and plane indices of the simulated grain boundary; (*b*) Atomistic view of the grain boundary plane at 0 K. Atom colors correspond to local atomic symmetry.

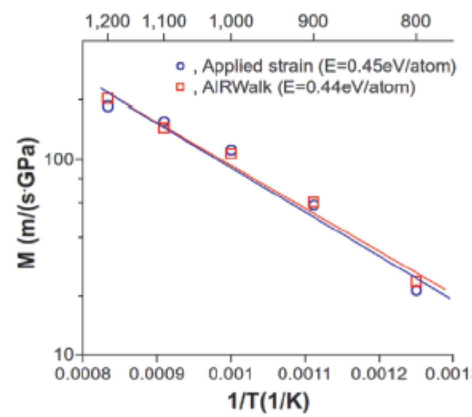
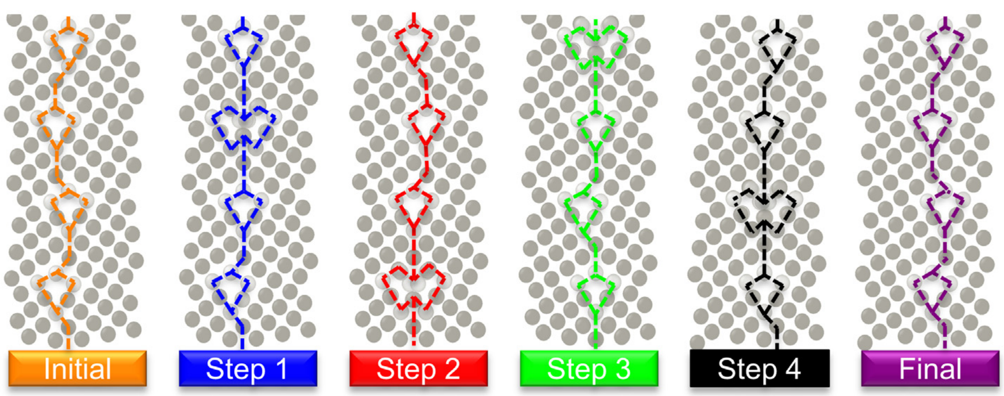


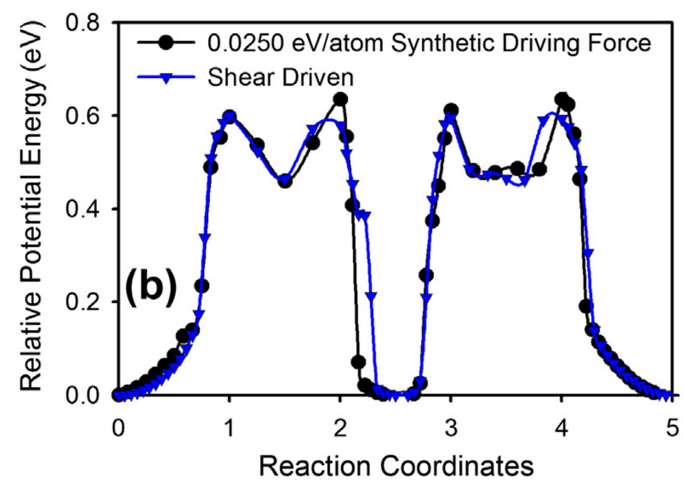
Figure 6. Temperature dependence of the GB mobility.

Mendelez, Deng, Schuh, Srolovitz, MSMSE 21 (2013) 045017

Computational Detail: Test shows that synthetic driving force has minimal effect on barriers

- Nudged elastic band calculations of the barriers for the motion of a large-angle [0 0 1] symmetric tilt boundary
  - Synthetic driving force
  - Shear driving force
- The path associated with the boundary motion and the associated barriers are very close





Coleman, Spearot, Foiles, Comp. Mat. Sci. 86 (2014) 38-42

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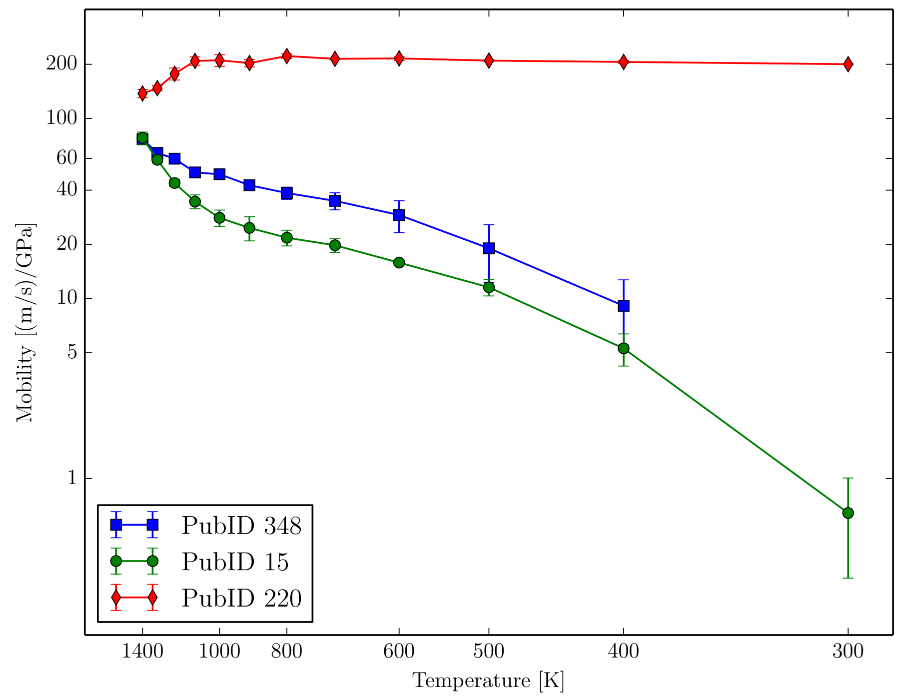
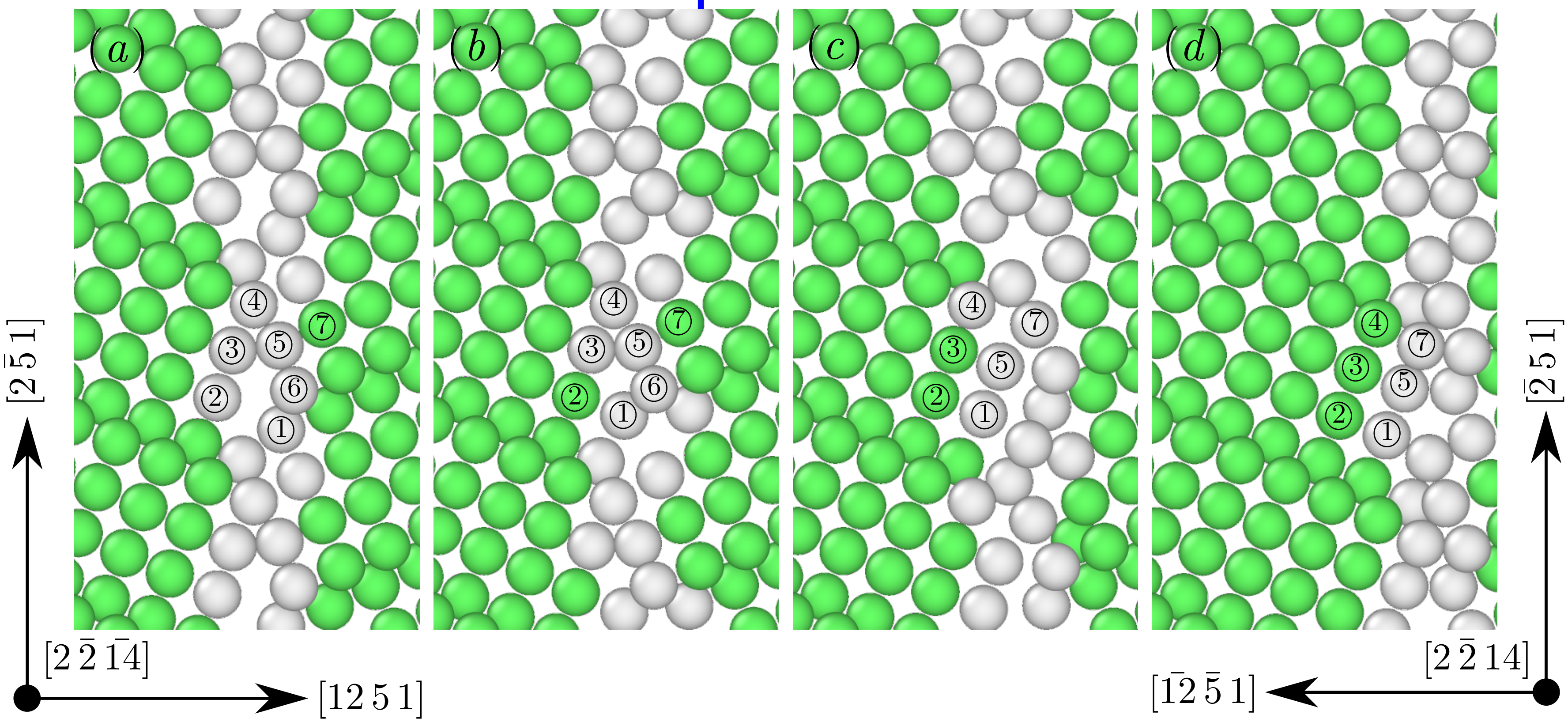
Case studies of boundary motion mechanisms

- Though Arrhenius behavior of the mobility with temperature is the conventional wisdom, the survey revealed existence of boundaries which defy this assumption
- Three grain boundaries with different temperature dependence of mobility
  - Roughly Arrhenius
    - Σ15 (12 5 1)/(12 5 -1)
    - "Pub ID 348"
  - Arrhenius with curvature
    - Σ5 (2 1 1)/(2 1 1)
    - "Pub ID 15"
  - Anti-thermal
    - Σ7 (12 3 1)/(9 8 3)
    - "Pub ID 220"

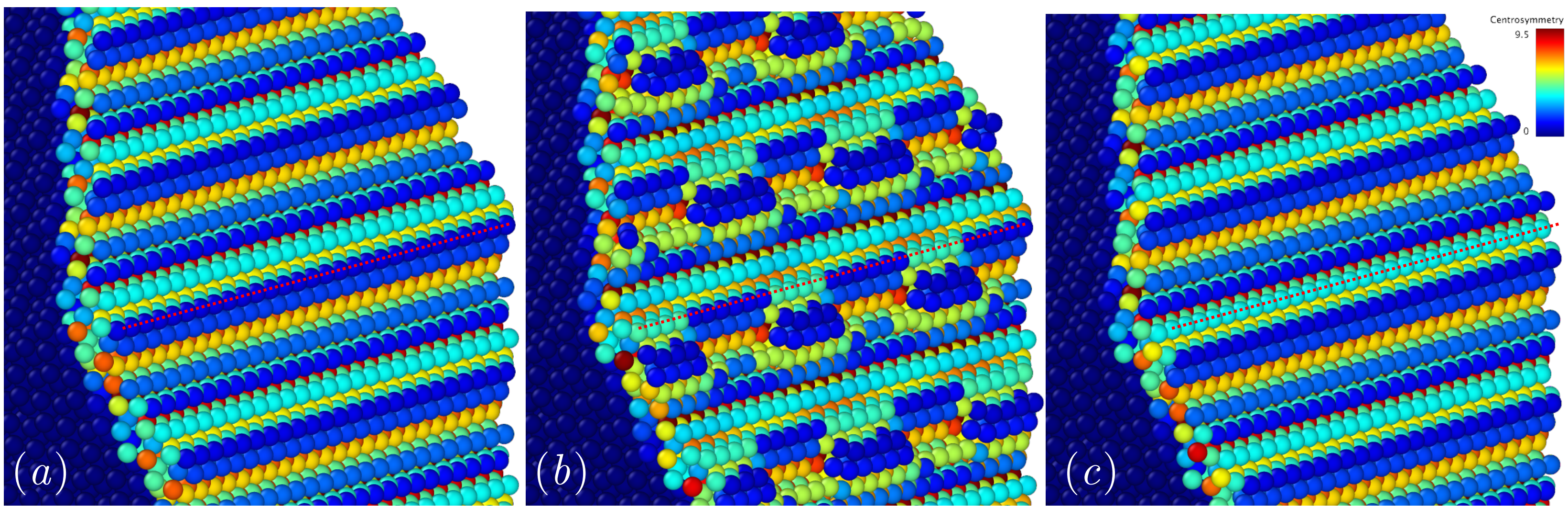


Arrhenius boundary (#348) moves by a complex shuffle



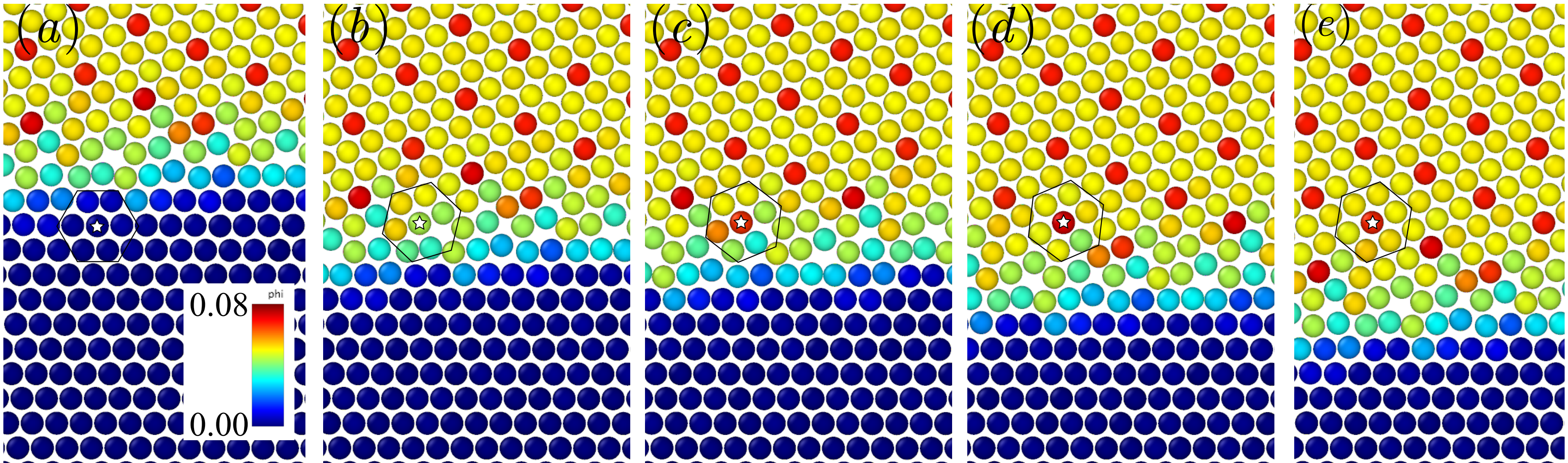
- Color coding: FCC, Other
- Note that atom “6” moves normal to the plane and out of the projected slice

Athermal boundary appears to move by a step process



- Coloring by centro-symmetry parameter
  - Red dashed line is a fiduciary marker

Athermal Σ7 boundary moves by the local rotation of atoms in the common (111) plane



- Coloring by microrotation
  - Star is a fiduciary marker
- Note that Σ7 boundaries have a common (111) plane
  - Motion occurs by rotation of an array of hexagons
- This mechanism may explain the observed high mobility of Σ7 boundaries

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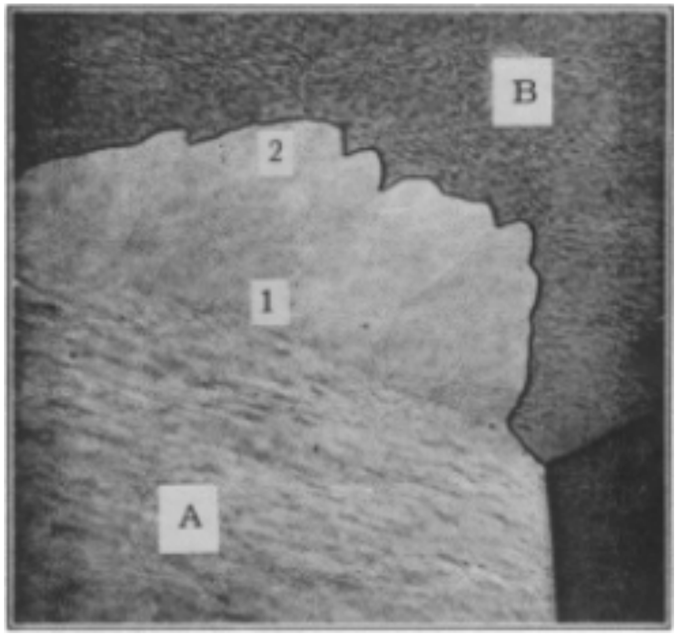
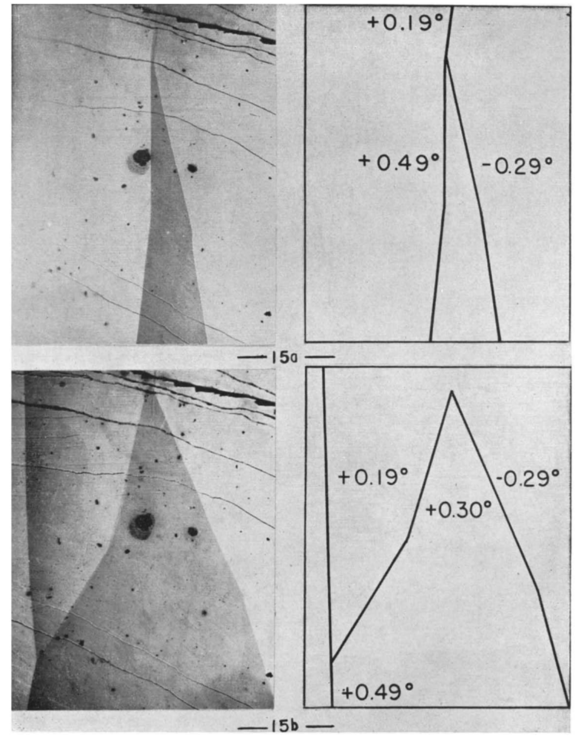
Mechanically induced grain growth – over a wide temperature range - has been recognized for decades



Plastic strain-induced boundary migration in deformed Al, observed during annealing at 350°C.

- Driving force is direct removal of stored dislocations by boundary sweeping.

Beck and Sperry, *J. Appl. Phys.* **21** (1950) 150.



Elastic stress-induced, reversible low-angle grain boundary migration in Zn bicrystals at -196°C and 375°C.

- Driving force is relief of elastic stress via grain boundary dislocation motion.

Bainbridge, Li, and Edwards, *Acta Metall.* **2** (1954) 322.

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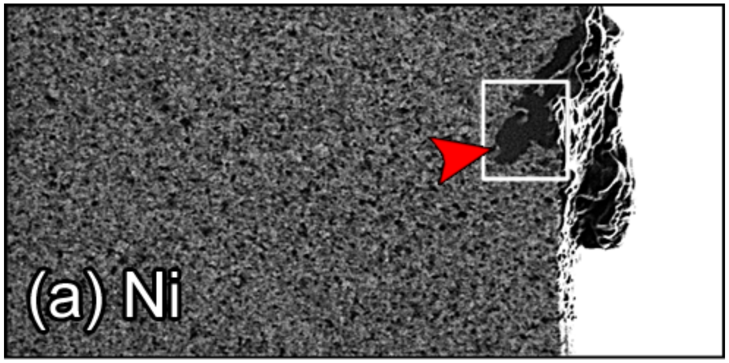
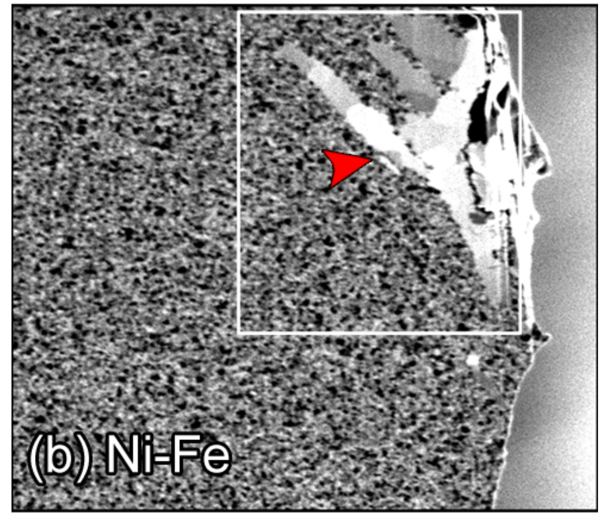
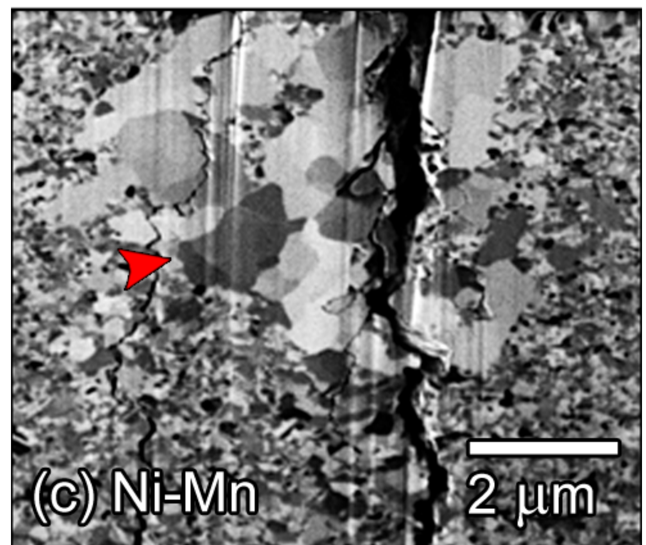
Mechanically-induced grain growth limits the fatigue life of nanocrystalline metals.



- During fatigue tests of nanocrystalline alloys, failure is always observed to initiate at colonies of very large grains.
- These abnormal grains develop during fatigue testing.
  - Room temperature
  - Nominally elastic
  - High Schmid factor grains
- In the absence of large grains, the material does not fail.

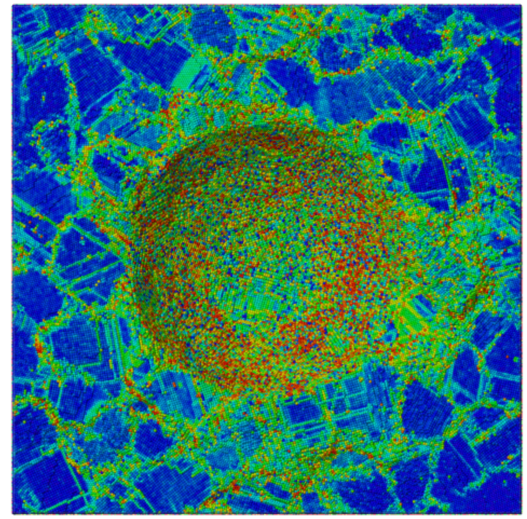




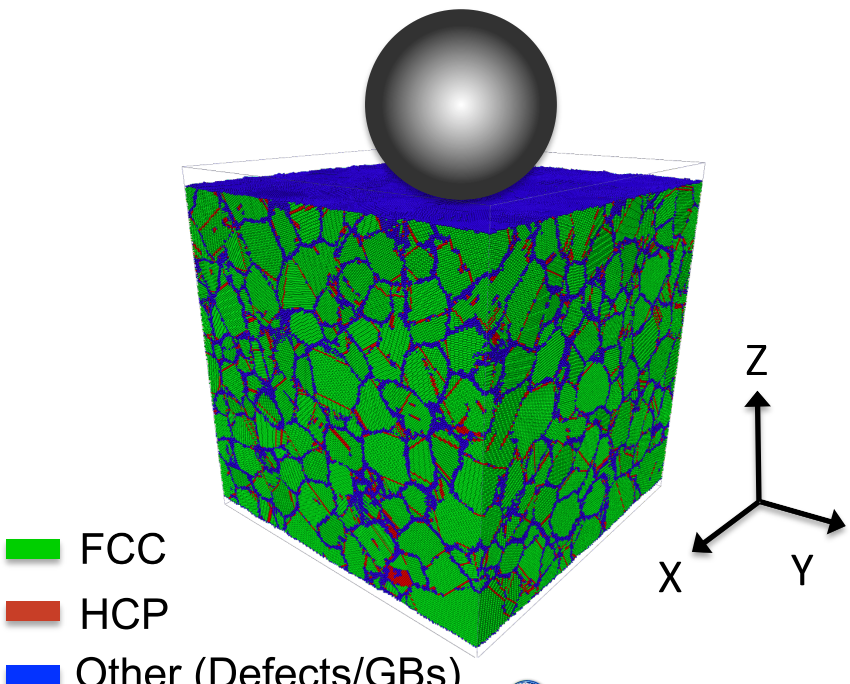
Boyce and Padilla, *Metall. Mater. Trans A* **42A** (2011) 1793.

Computational Methodology – Initial State

- Thin film (X/Y periodicity, free surface in Z)
- 3D Voronoi tessellation followed by thermal anneal
  - Pre-anneal: ~4nm grain size
  - Random orientations
  - $(53\ \mathrm{nm})^3$ containing about 13 million atoms
- Thermal equilibration at 1175 K (0.75 T_m) for 0.2 ns
  - **Thermal grain growth yields**
    - **half the number of initial grains**
    - **equilibrates triple junctions**
- Ni EAM potential (Foiles et al. 2006)



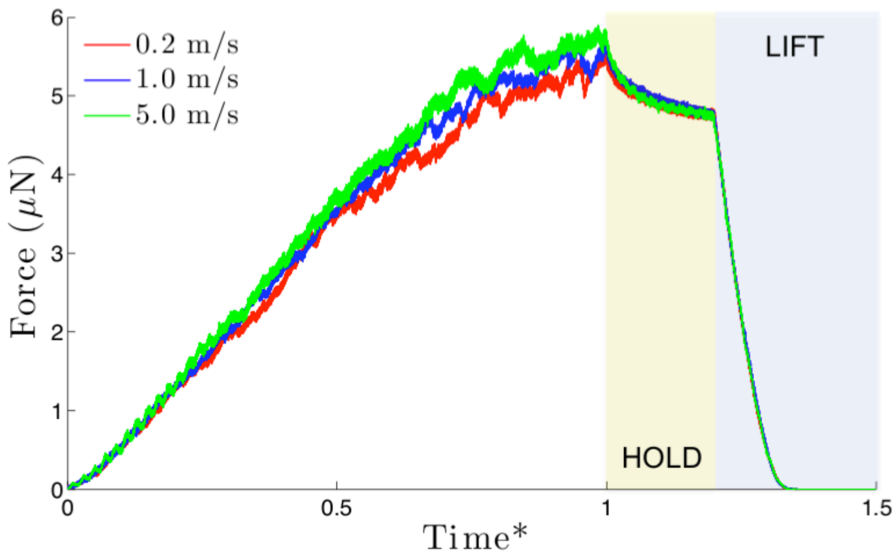
- R=15 nm spherical indenter
- Indenter modeled by repulsive potential

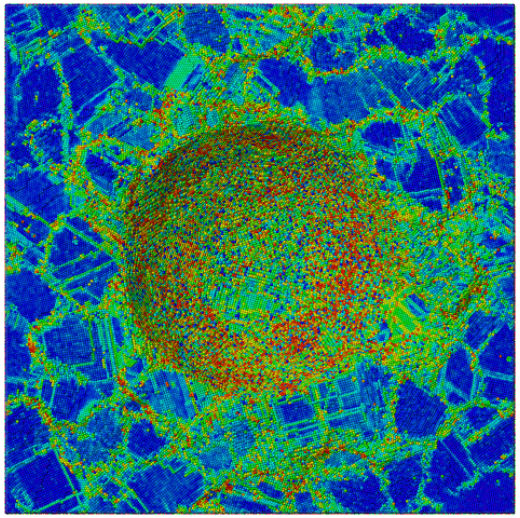


- **Constant velocity indentation:**
  - **0.2 m/s, 1.0 m/s, and 5.0 m/s**
- **Three phases:**
  - **Indentation**
  - **Hold**
  - **Withdrawal of indenter.**

Tucker and Foiles, Materials Science and Engineering A 571, 207 (2013)

Computational Methodology – Indentation History



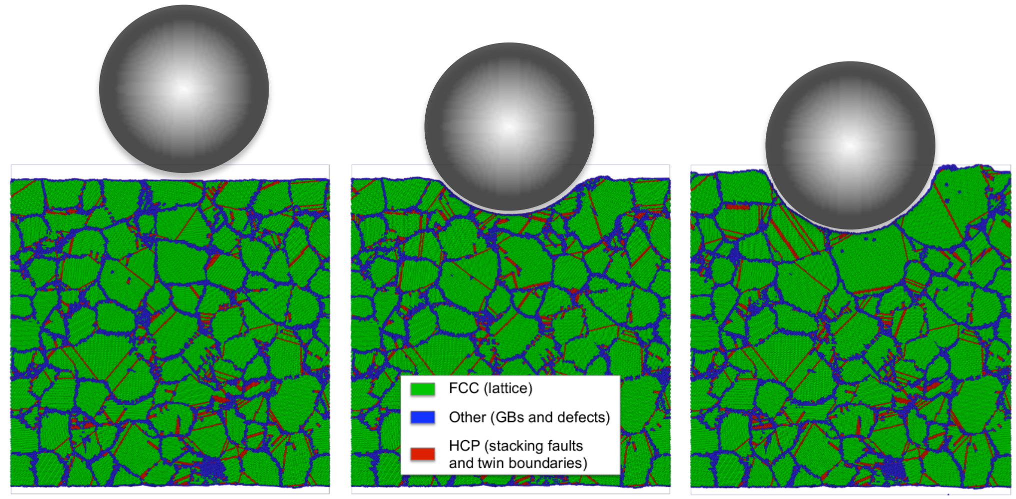


- R=15 nm spherical indenter
- Indenter modeled by repulsive potential

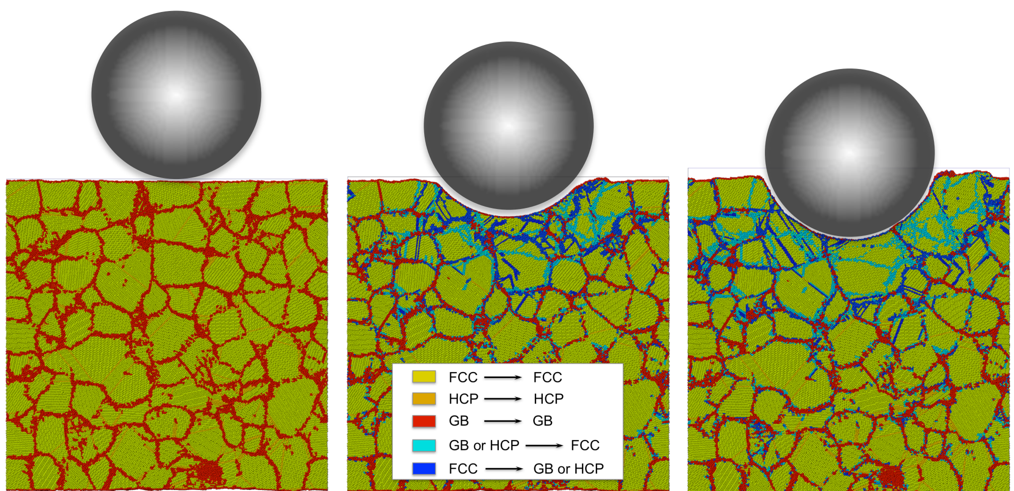
- Indentation force shows a modest rate dependence during indentation
  - Force largest for fastest indent
- The normalized relaxation rate during the 'hold' and 'lift' phases is approximately identical for all three indentation rates.
  - During the hold and relaxation, the force only depends on indenter depth

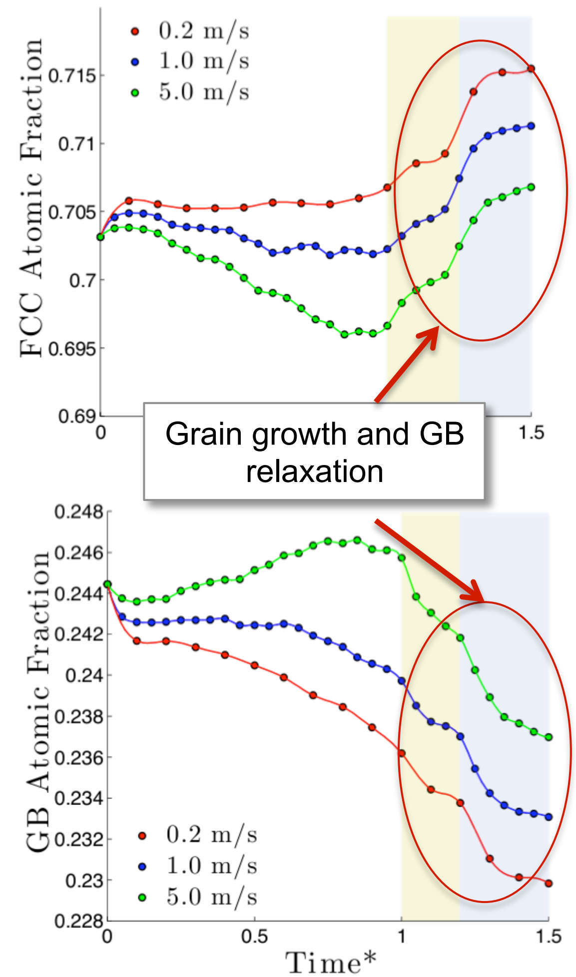
- **Constant velocity indentation:**
  - **0.2 m/s, 1.0 m/s, and 5.0 m/s**
- **Three phases:**
  - **Indentation**
  - **Hold**
  - **Withdrawal of indenter.**

Analysis can track evolution of grain boundary area and the motion of grains



Comparison of initial and final states shows the elimination of grain boundaries near the indenter



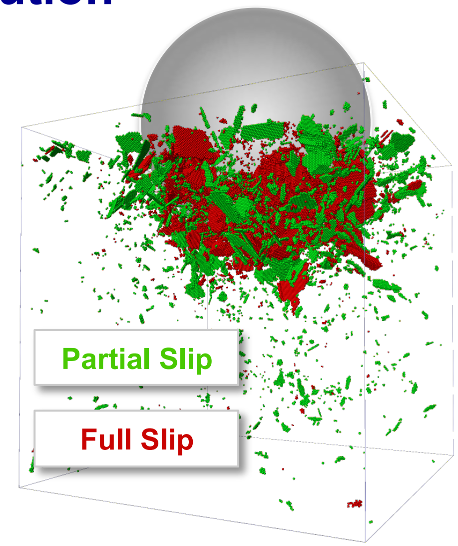


Significant rate dependence in the evolution of grain boundary area

Tucker and Foiles (2012), Mat. Sci. Eng. A 571, 207 (2013)

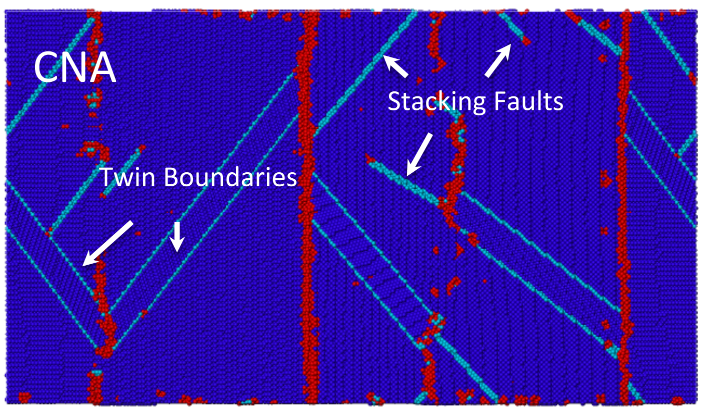
Combination of analysis methods reveals the dislocation and twin boundary evolution

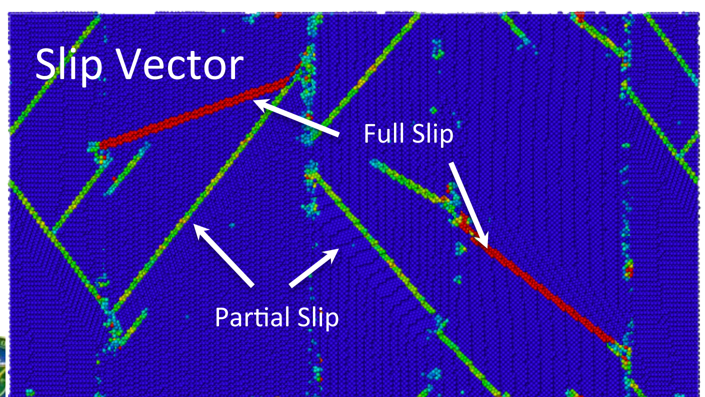
- Utilize neighbor lists and computed metrics to determine atomic slip and local neighborhood.

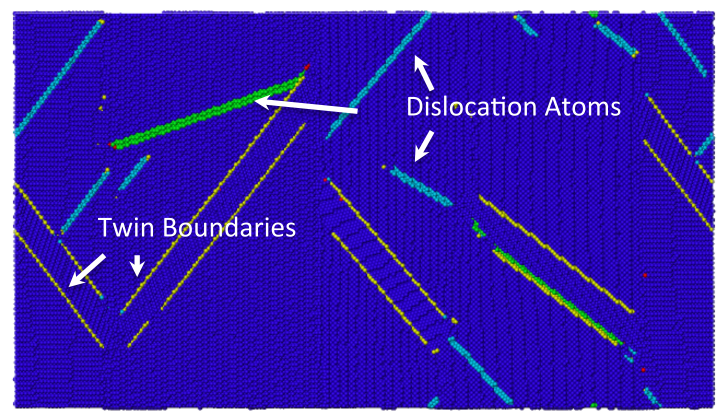


Slip Vector $$s^{\alpha} = -\frac{1}{n_s}\sum_{\beta \neq \alpha}^{n}(x^{\alpha\beta} - X^{\alpha\beta})$$

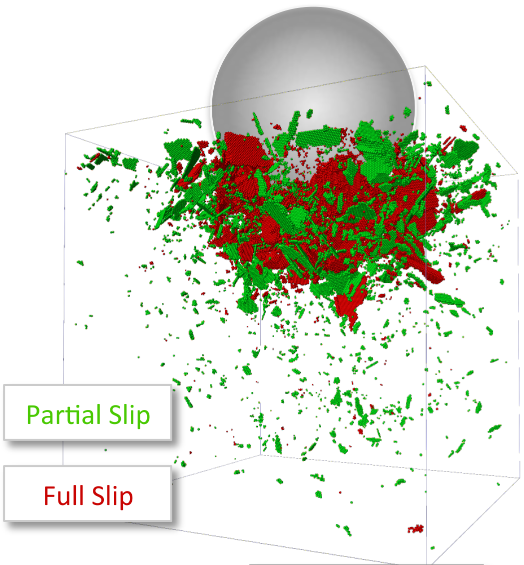
- Assign non-GB atoms integer values for deformation mechanism group
  - Dislocations (Partial/Full)
  - TBs



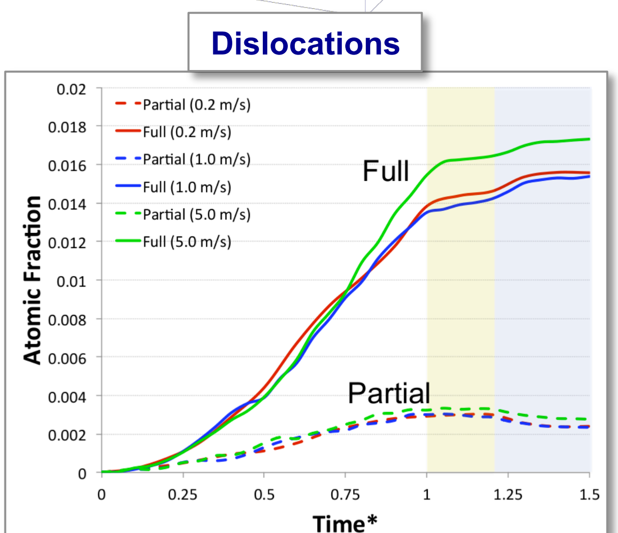


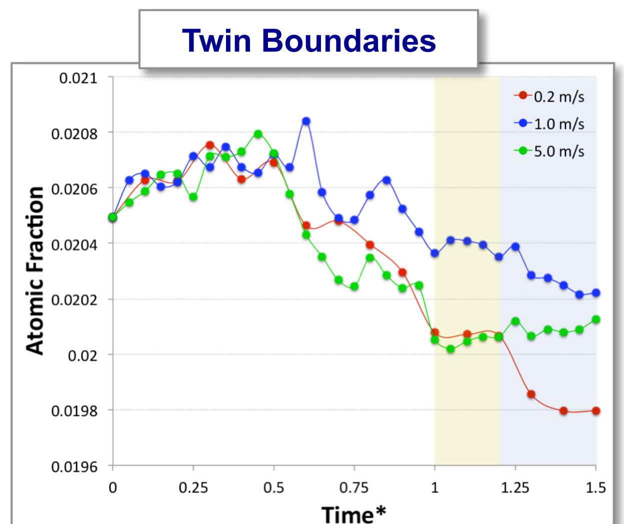


Both dislocation slip and twin boundary density evolution occur during the indentation

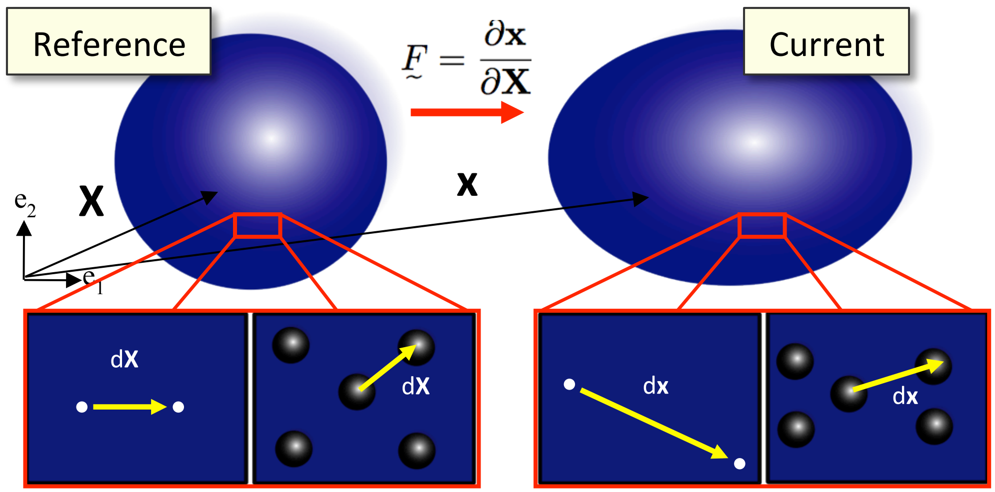


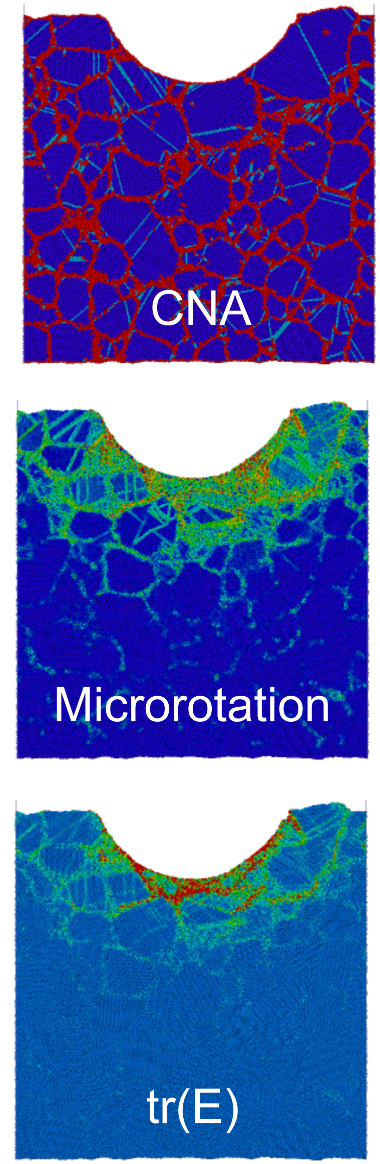
How do we quantify the relative importance of these mechanisms?





Microscale Kinematic Metric Formulation reveals local deformation modes





Deformation Gradient

$$F = \frac{\partial x}{\partial X} \rightarrow \sum_{\beta=1}^{n} \left(x_i^{\alpha\beta} X_M^{\alpha\beta} - F_{iI}^{\alpha} X_I^{\alpha\beta} X_M^{\alpha\beta}\right) = 0 \rightarrow \boxed{F_{iI}^{\alpha} = \omega_{iM}^{\alpha} \left(\eta^{\alpha}\right)_{MI}^{-1}}$$

where $\omega_{iM}^{\alpha} = \sum_{\beta=1}^{n} x_i^{\alpha\beta} X_M^{\alpha\beta}$

and $\eta_{iM}^{\alpha} = \sum_{\beta=1}^{n} X_I^{\alpha\beta} X_M^{\alpha\beta}$

* Zimmerman *et al.*, IJSS (2009)

Microrotation

$$\underset{\sim}{F} = \underset{\sim}{R}\underset{\sim}{U} \text{ where } \underset{\sim}{R} = \underset{\sim}{R}_{sym} + \underset{\sim}{R}_{skew} \rightarrow \underset{\sim}{R}_{skew} = \frac{1}{2}(\underset{\sim}{R} - \underset{\sim}{R}^T) \rightarrow \boxed{\phi_k = -\frac{1}{2} \varepsilon_{ijk} \left(R_{skew}\right)_{ij}}$$

* Tucker *et al.*, MSMSE (2010)

Green Strain

$$\boxed{\mathbf{E} = \frac{1}{2}(\mathbf{F}^{\mathrm{T}} \cdot \mathbf{F} - \mathbf{I})} \quad \text{and} \quad I_1(E) = tr(E)$$

* Tucker *et al.*, JMPS (2012)

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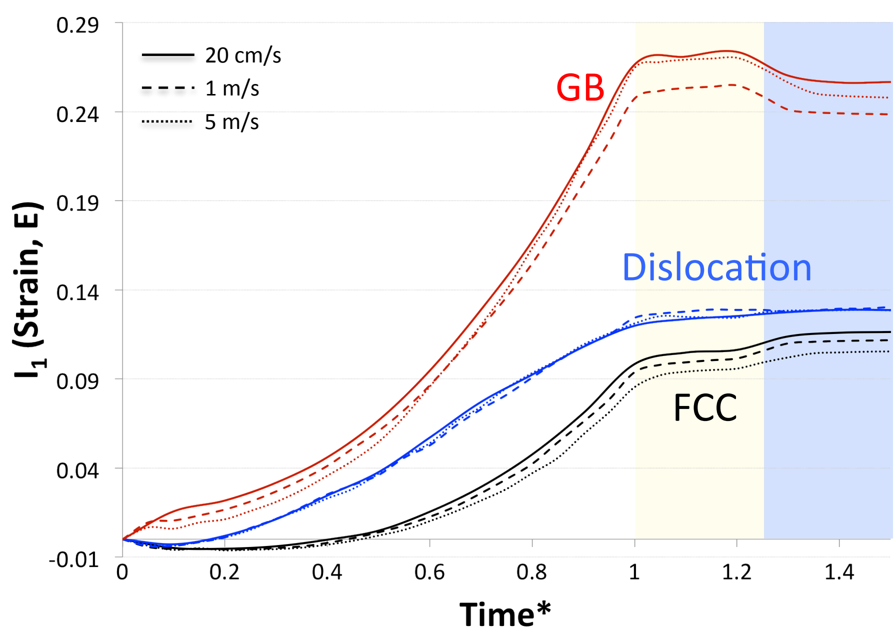
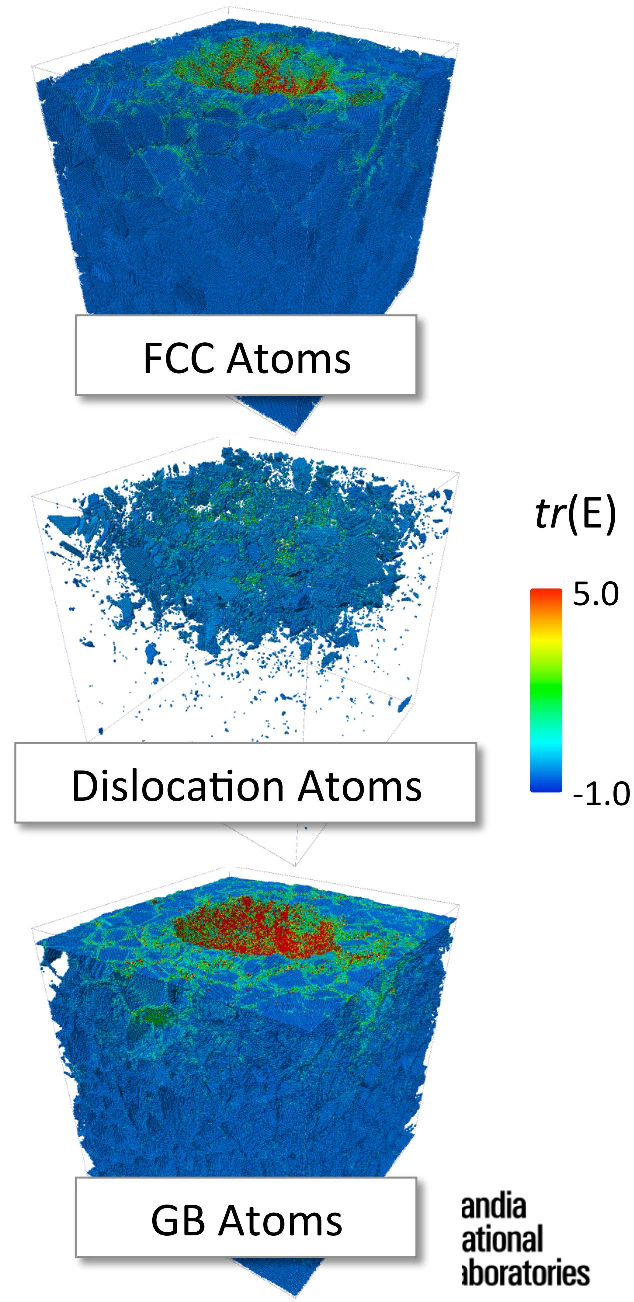
Strain Accommodation is Localized to Interfaces and Dislocations

Deformation Gradient Tensor $F^{\alpha}_{iI} = \omega^{\alpha}_{iM}(\eta^{\alpha})^{-1}_{MI}$

Green Strain Tensor $\boldsymbol{E} = \frac{1}{2}(\boldsymbol{F^T} \cdot \boldsymbol{F} - \boldsymbol{I})$

- *tr*(E) from all atoms in a phase (e.g. FCC, GB, Dislocation) added together.
- Strain accommodation from Interface plasticity is substantial
  - Some relaxation during release
- Dislocation plasticity is roughly rate-independent and does not relax during release





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Summary

- MD simulations have been used to explore the variation of properties of planar grain boundaries over geometry and temperature
  - Trends in properties are being quantified
  - Significant numbers of boundaries don't conform to conventional wisdom
- MD simulations of nanocrystalline deformation reveal the interplay between conventional dislocation mechanisms and boundary deformation
  - Significant simulation post-processing and concepts from continuum mechanics required to quantify the relative importance of different mechanisms
- Now you must answer the most important question of the day
  - ***RED or GREEN!***

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